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[illegible]e Treatment of the Waste Water of Urea Fertilizer Plant with a Combined *Process of Advanced* *[illegible]idation* and Microalge *Chlorella pyrenoidosa,* *[illegible]nochloropsis* sp. and *Pseudomonas fluorescens* Bacteria

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[illegible] – [illegible] activity of urea fertilizer industry with potential [illegible]mental pollution is the activity of wastewater [illegible] waters. The main by product of the fertilizer urea [illegible] containing mostly liquid ammonia. According to [illegible] ministry of Environmental Affairs No. 122 of the [illegible] the Decree of the Governoor of South Sumatra [illegible] of the year 2005, the maximum pollution load for [illegible] industry is liquid ammonia level of 0.75 kg/ton (50 [illegible] 6.0 – 9.0. The treatment of the wastewater of urea [illegible] done by means of combining chemical and bilogical [illegible] technology of wastewater treatment by advanced [illegible] using strong oxidizing agents. The process of [illegible] combined with or followed by biological processes [illegible]organisms such as microglae *Chlorella pyrenoidosa,* [illegible] And the *Pseudomonas fluorescens* bacteria. The [illegible] study on the treatment of the wastewater of the urea [illegible] using Fenton reagent and advanced oxidation [illegible] follows: the capacity to degrade NH_3 is 95% and [illegible] ratio of 1:10 and the use *Pseudomonas fluorescens* [illegible] nitrate and nitrite as much as 92.63% microalgae [illegible] as much as 99% and *Nannochloropsis sp.* as [illegible]

[illegible]—[illegible]anced Oxidation Process (AOP), Chlorella [illegible]nochloropsis sp, Pseudomonas fluorescens

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I. INTRODUCTION

THERE are six urea fertilizer plants in Indonesia whose wastewater is characterized with high levels of urea and ammonia-nitrogen. The treatment of wastewater with high levels of urea and ammonia-nitrogen is one of the problems faced by urea fertilizer plants in Indonesia. Although the waste water of urea fertilizer plants is not considered as hazardous materials compound, it may cause serious damage to the ecosystem of water bodies. Industrial activities of urea fertilizer plants with the potential impact of causing environmental pollution is the disposal activities of wastewater into the waters. The efforts to improve wastewater treatment by separating ammonia have been done by using variety of methods such as: *Ion Exchange* [20], *breakpoint chlorination* [12], aerobic-nitrification and *anaerobic-denitrification electron* [1], *water striping* [29], *fluidized-bed reactor* [15], *anammox* (*Anaerobic Ammonium Oxidation*) [21-23], *combined nitrification-denitrification and microalgae* [25], *membrane reactors* (*MBR*) [28]. These ammonia separation methods have limitations, such as not being able to reduce the amount of ammonia up to safe concentration level, needing huge cost and the application of some of these methods in practice still encounters obstacles. This constraint is mainly due to the specific capacity of NH_3-N removal is still so low that the output process is still higher than the quality standards that have been set.

... according to [10] biological waste treatment ... microbes) will not run optimally or will be impaired ... chemical compounds are found in the wastewater ... affect the performance of a waste treatment facility. In ... the treatment of the wastewater of the urea fertilizer ... carried out by combining chemical and biological ... The chemical processing technology applied in this ... Advanced Oxidation Proces (AOP) or advanced ... processes of wastewater treatment which is a ... processing techlogy with advanced oxidation ... using strong oxidator. This process of oxidation can ... combined or followed by biological processes involving ... organisms such as microalgae *Chlorella pyrenoidosa*, ... sp. and the bacterium *Pseudomonas* ...

II. Materials And Methods

Material and Tools

... tools used in this study are volumetric flask, pH meter, ... pipette, Spectrophotometer, scales, aerator, ... plastic tubing measures ¾ diameters, ... microscope, fluorescent lamp and the culture ... drill, Petri dishes, transparent millimeter paper. The ingredients needed are water, distilled water, Nessler ... ammonia derived from Wetland area of nitrogen ... *C. pyrenoidosa* and *Nannochloropsis* sp, ... from pure cultures in the uncontaminated ... or other organisms. Water used is ... a mixture of sea water and fresh water) as the main ... of *Nannocloropsis sp* with 3% salinity, while ... *C. pyrenoidosa* using fresh water, pH 8 - 9.5, ... of 25-30 ° C. Bacteria P. fluorescens seeds ... pure cultures in the uncontaminated condition. ... composition are protease peptone 10 g, ... $MgSO_4.7H_2O$ 0, 75 g, glycerol 7, 5 ml, drilled ... variable measurement including pH, density, ... nitrogen, H2O2, FeSO4.7H2O, Measurement ... pH, NH3-N, urea, nitrate, nitrite, TKN, COD, ...

... Research

... from a pool emergency tube inserted into the ... masukkeumpan wastewater is analyzed ... nitrate, nitrite, TKN, pH, COD, TSS)

... inserted control samples (waste water) into ... of 5.000 (reagent tubes). The tube serves as a ... to react with the fertilizer plant wastewater ... and various comparisons are $FeSO_4$: H_2O_2 1: ... 1: 8, 1: 10.

... magnetic stirrer at 100 rpm stirring speed setting ... minutes, and then the samples were taken after ... Then the waste water from the feed tube in ... reagent (NH_3-N, urea, nitrate, nitrite,, TKN, ... TSS).

- Waste water from in vitro reagent, flowed into aguarium / botolaerasi (aguarium microalgae, bacteria, bacteria + microalgae), the aerobic process. Later on leave for 7-9 days, because the growth of microorganisms reach stationary phase at 4-6 days. So that microorganisms (bacteria and microalgae) can decompose organic substances contained in waste water
- Water processed, the water that comes from aguarium / aeration bottle, then analyzed (NH_3-N, urea, nitrate, nitrite, TKN, pH, COD, TSS), which is useful to know the quality of the waste water from some of the previous process .

C. Circuit Research Tool

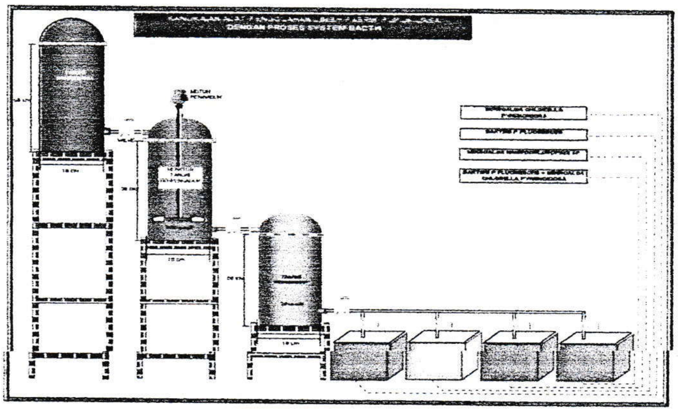

Fig 1. Circuit Research Tools

III. Results And Discussion

A. Fenton Reagent Result

The treatment of the wastewater of urea fertilizer plant using Fenton reagent of various concentrations and comparisons of $FeSO_4$ and H_2O_2 causes a decline in NH_3-N, Urea, COD, TKN, and TSS, but an increase in levels of pH, nitrate and nitrite as presented in Table 1 . The results of the study show a precipitate $Fe(OH)_3$ which is reddish brown, because the wastewater of the urea fertilizer plant used is that with the acidity (pH) levels of more than 6 [9]. The decline in the value of NH_3-N and urea in the results of the study presented in Table 1, is assumed to decompose to form ions and gases, such as nitrate and nitrite molecule, or nitrogen monoxide. This is in accordance with the findings of [27] , that the levels of nitrate and nitrite increase. The levels of COD, TSS decrease, indicating that Fenton oxidation can degrade the value of COD and TSS. According to [2], Fenton reagent as one of the advanced oxidation processes (*Advanced Oxidation Process / AOPs*) is expected to destroy organic and inorganic pollutants, eliminate color and COD.

TABLE I.
[illegible] TREATMENT WASTEWATER UREA FERTILIZER [illegible] REAGENT CONCENTRATION 2500, 2000 AND 1500 PPM

[illegible]	Ratio between $FeSO_4$ (gram) : H_2O_2 (ml)				
[illegible]	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
[illegible]	8,4	8,6	8,9	9,1	9,7
[illegible]	302,25	237,52	298,59	262,27	157,50
[illegible]	2808	2590	2341	2335	1941
[illegible]	16,50	19,23	23,12	59,75	113,85
[illegible]	2,365	3,21	3,725	4,346	7,03
[illegible]	1602,29	1596,18	1391,15	1305,36	1143,30
[illegible]	5	5	4	2	2
[illegible]	1,98	1,55	2,20	2,35	2,23
[illegible]	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
[illegible]	8,2	8,5	8,7	8,8	9,3
[illegible]	119,47	73,75	11,15	4,625	1,612
[illegible]	1523	1478	1405	1119	665
[illegible]	15	18,17	22,89	38,29	86,39
[illegible]	1,632	2,624	2,579	3,132	4,97
[illegible]	715,5	693,63	657,8	514	307,512
[illegible]	5	3	2	2	2
[illegible]	1,56	1,78	2,23	2,12	2,88
[illegible]	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
[illegible]	8,0	8,2	8,3	8,5	9,0
[illegible]	17,50	4,25	4,75	0,12	0,025
[illegible]	2057	605	759	439	32
[illegible]	15,60	17,12	19,65	29,65	52,34
[illegible]	1,156	2,611	2,556	3,046	3,48
[illegible]	505,72	417,05	342	201	14,743
[illegible]	3	3	2	2	2
[illegible]	1,63	1,33	2,0	2,27	2,87

[illegible] reagent and Microalgae Chlorella [illegible] and Nannochloropsis sp.

[illegible] on the treatment of wastewater of the urea [illegible] using Fenton reagent and microalgae Chlorella [illegible] and Nannochloropsis is a further research [illegible] the results Fenton reagent which indicates the [illegible] of increasing levels of nitrate and nitrite at a [illegible] of 2500 ppm, 2000 ppm and 1500 ppm . This [illegible] of NH_3-N and urea is expected to be absorbed by [illegible] microalgae Chlorella pyrenoidosa and [illegible] sp. as nutrients. The results of the study [illegible] Tables 2 and 3 show no changes in pH value [illegible] by using microalgae Chlorella [illegible] and Nannochloropsis sp, no increase or decrease [illegible] the pH value is relatively stable during the [illegible] to [5], the pH did not increase due to the [illegible] natural buffer system in the form of dissolved [illegible] in the culture medium. The dissolved CO2 [illegible] in the medium will become carbonic acid which [illegible] into ions. The results of the study presented in [illegible] 3 show the increase of COD and TSS values [illegible] there is an accumulation of organic materials [illegible] resulting from the growth of microalgae [illegible] and *Nannochloropsis sp.*

TABLE II.
RESULT ANALYSIS TREATMENT WASTEWATER UREA FERTILIZER PLANTS FENTON REAGENT AND MICROALGAE *CHLORELLA PYRENOIDOSA* CONCENTRATION 2500, 2000 AND 1500 PPM

Parameter	Initial Ratio Analysis $FeSO_4$ (gram) : H_2O_2 (ml)				
Concentration (2500 ppm)	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
pH	8,4	8,6	8,9	9,1	9,7
NH_3-N	135	35	103,75	124	129,11
Urea	1914	1803	1685	2075	1816
Nitrate	0,032	1,87	9,37	44,75	99,69
Nitrite	0,393	0,076	0,346	2,546	6,08
TKN	1028,2	763,07	892,08	963,005	933,243
COD	130	105	119	138	110
TSS	31	31	31	37	37
Concentration (2000 ppm)	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
pH	8,2	8,5	8,7	8,7	9,3
NH_3-N	0,25	0,15	0,013	0,013	0,0
Urea	695	713	702	702	529
Nitrate	0,010	0,21	6,35	6,35	76,8
Nitrite	0,056	0,039	0,056	0,056	3,39
TKN	333,65	239,55	327,613	327,613	181,553
COD	140	115	121	121	120
TSS	35	35	35	35	59
Concentration (1500 ppm)	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
pH	8,0	8,5	8,3	8,5	9,0
NH_3-N	0,034	0,15	0,001	0,0	0,0
Urea	157,05	713	37	32,45	0,0
Nitrate	0,034	0,21	3,9	16,95	39,89
Nitrite	0,037	0,039	0,037	0,245	2,7
TKN	73,324	239,55	17,266	15,143	0,0
COD	154	115	150	170	134
TSS	43	35	42	45	40

High COD level indicates an organic waste pollution. The inorganic materials found are in the form of clay and sand, and the organic matters are in the form of the remains of plants and other biological solids such as algae cells, bacteria and so forth [18]. Based on the results of the study, it can be concluded that the rise of TSS levels is due to the growth of microalgae *Chlorella pyrenoidosa* and *Nannochloropsis* sp.

TABLE III.
RESULT ANALYSIS TREATMENT WASTEWATER UREA FERTILIZER PLANTS FENTON REAGENT AND MICROALGAE *NANNCHLOROPSIS* SP. 2500, 2000 AND 1500 PPM

Parameter	Initial Ratio Analysis $FeSO_4$ (gram) : H_2O_2 (ml)				
Concentration (2500 ppm)	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
pH	8,4	8,6	8,9	9,1	9,7
NH_3-N	162,25	98	119,2	174	131
Urea	2195	1807	1884	2132	1895
Nitrate	2,63	3,50	13,45	47,75	109,69
Nitrite	0,921	0,90	1,937	3,345	6,84
TKN	1021,95	924,2	775	912,60	1031,36
COD	178	170	166	234	159
TSS	50	40	47	50	53
Concentration (2000 ppm)	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
pH	8,4	8,5	8,7	8,8	9,3
NH_3-N	0,425	0,075	0,073	0,078	0,0
Urea	925	634	930	923	543,34
Nitrate	2,28	1,65	9,5	27,31	71,8
Nitrite	0,567	0,870	1,30	1,30	2,03
TKN	425,92	383,715	335	370,144	31,21
COD	180	180	181	138	162
TSS	52	52	52	52	52
Concentration (1500 ppm)	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
pH	8,0	8,2	8,3	8,5	9,0
NH_3-N	0,662	0,0	0,002	0,0	0,0
Urea	326	296	138	64,01	1,56
Nitrate	1,84	1,34	6,36	12,93	31,14
Nitrite	0,470	0,774	0,567	1,10	1,67
TKN	196,02	103,96	64,402	29,835	0,728
COD	205	205	167	197	177
TSS	52	52	52	52	69

[illegible] and *Pseudomonas fluorescens*

[illegible] of waste water using bacteria *Pseudomonas* [illegible] research, in which waste water of urea [illegible] is treated using Fenton reagent of various [illegible] from 2500 ppm, 2000 ppm and 1500 [illegible] indicate that it still contains high levels of [illegible] increase in the levels of nitrate and nitrite. [illegible] study presented on Table 4 show changes in [illegible] (pH), namely a decline. The reason is the [illegible] carried out by the bacteria that produce [illegible] of CO_2 in the water will shift the carbonate [illegible] right so that it will lower the pH value. The [illegible] equilibrium reaction of the carbonate:

$$[illegible] \longleftrightarrow OH^{+} + HCO_3 \quad (1)$$

[illegible] presence of CO_2, the reaction equilibrium will shift [illegible] the H^+ ions that will cause a decrease of the [illegible]. This is in accordance with the opinion of [16], [illegible] an increase in CO_2 will lower the pH value of [illegible] the increase in maintenance time, the amount [illegible] increase which will further decrease the pH [illegible] media. The CO_2 in the media is also allegedly [illegible] decomposition of organic matter and respiration [illegible]. According to [26], the bacteria will use organic [illegible] energy source, in correlation with the nitrogen to [illegible] protein synthesis in order to produce new cell [illegible] the addition of carbonic materials, the bacteria [illegible] nitrogen contained in the culture so as to reduce the [illegible] of inorganic nitrogen (ammonia) which is toxic [illegible].

TABLE IV.
[illegible] TREATMENT WASTEWATER UREA FERTILIZER [illegible] REAGENT AND *PSEUDOMONAS FLUORESCENS* [illegible] CONCENTRATION 2500, 2000 AND 1500 PPM

[illegible]	Initial	Ratio Analysis FeSO₄ (gram) : H_2O_2 (ml)			
[illegible]	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
[illegible]	8,1	8,4	8,7	8,9	9,7
[illegible]	76,5	55	76,25	107,25	119,56
[illegible]	[illegible]	1080	1495	1479	1770,09
[illegible]	1,20	0,98	0,67	32,78	92,45
[illegible]	[illegible]	0,076	0,043	1,087	6,98
[illegible]	[illegible]	727,35	697,91	727,44	928,60
[illegible]	13	11	18	17	15
[illegible]	10	12	11	13	10
[illegible]	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
[illegible]	7,8	8,2	8,3	8,7	9,2
[illegible]	0,125	0,08	0,007	0,0	0,0
[illegible]	[illegible]	306	693,67	583,07	300,4
[illegible]	[illegible]	0,34	0,56	22,08	69,90
[illegible]	[illegible]	0,034	0,023	1,024	3,053
[illegible]	[illegible]	232,84	323,782	272,09	12,290
[illegible]	15	15	15	19	14
[illegible]	14	15	15	16	14
[illegible]	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
[illegible]	7,7	7,8	8,0	8,3	8,6
[illegible]	0,075	0,003	0,0	0,0	0,0
[illegible]	248	15	145,78	1.056	0.0
[illegible]	[illegible]	0,26	0,91	17,45	69,90
[illegible]	0,064	0,019	0,011	0,019	0,12
[illegible]	68,95	6.093	0,207	0,482	0,0
[illegible]	20	15	23	19	25
[illegible]	15	17	15	17	15

The addition of carbonaceous material is proven to be [illegible] of reducing inorganic nitrogen [2]. Bacterial growth is [illegible] the balance of nutrients in the water. Therefore, the dynamics of bacteria population are closely related with the availability of nutrients [10]. The data in Table 4, show that the levels of nitrate and nitrite decrease. The decrease is due to the fact that the nitrate and nitrite formed are sufficient for bacterial nutrients that will stimulate the growth of bacteria and the increase of bacterial biomass Although in low concentrations, nitrite is toxic to fish and other aquatic organisms [13]. Nitrite compounds in fish will be bound in blood that will form methaemoglobin ($Hb + NO_2 = Met\text{-}Hb$). The Met-Hb would interfere with the transport of oxygen to the tissues of fish that can cause fish to experience hypoxsia. The Met-Hb in the blood will cause the blood to become brown. Therefore, nitrite poisoning is also called brown blood disease [4]-[1]-[24]. The levels of Total Suspended Solid (TSS) and Chemical Oxygen Demand (COD) increase which indicates that there is accumulation of organic materials in the wastewater derived from *Pseudomonas fluorescens* bacteria growing in the waste water of urea fertilizer plants, which is in a state of non-toxic. Whereas the increase of TSS level is due to the occurrence of wastewater suspended solids which form residual components, floating materials, and suspended colloidal components. Suspended solids contain inorganic and organic materials. The inorganic materials are in the form of clay and sand, while the organic matters are in the form of the remains of plants and other biological solids such as algae cells, bacteria, and so forth [17].

D. Fenton Reagent, Microalgae Chlorella pyrenoidosa and Bacteria Pseudomonas fluorescens

In further processing of the treatment of the waste water of the urea fertilizer plants which contains high levels of ammonia and urea using Fenton reagent, it is expected that it will be continued with the use of a combination of microalgae *Chlorella pyrenoidosa* and bacteria *Pseudomonas fluorescens.* The microalgae and the bacteria will work together in degrading the waste water. The results of this study presented on Table 5. in general show that pH values tend to decrease with the increasing of maintenance time. This fluctuating value is allegedly due to the addition of the nitrification and denitrification bacteria into the maintenance media. The mechanism of bacterial nitrification and denitrification which can affect pH level can be described by the following equation [17].

Nitrification:

$$NH_4 + 2O_2 \longrightarrow NO_3 + 2H+ + H_2O \quad (2)$$

Denitrification :

$$NO_3 + H^+ \longrightarrow \tfrac{1}{2}\,(H_2O + N_2) + 5/2\,O_2 \quad (3)$$

Through this equation it can be seen that nitrifying bacteria in the process to convert NH_4^+ (ammonium) to NO_3^- (nitrate) produce H^+ ions that can make the pH of the maintenance medium drop. Since in a large-scale microalgae always associate with bacteria, the interaction between algae and bacteria will be able to purify river water. Metabolic activities of heterotrophic aerobic bacteria produce CO_2, NH_4, NO_3, PO_4^{3-}, and so on. The microalgae absorb those [illegible] produce organic matters, O_2, and H_2O. The [illegible] by *microalgae is used by heterotrophic* [illegible] for the reaction of nitrification and is used by [illegible] for denitrification. Through the process of [illegible] microalgae using CO_2 from aerobic bacteria [illegible] form cell protoplasm and release oxygen [illegible] - [19] - [22].

$$[illegible] + 4{,}5\ H_2O \xrightarrow{Light} C_5H_{14}O_3N + 8.75\ O_2 \quad (4)$$

TABLE V.
[illegible] TREATMENT WASTEWATER UREA FERTILIZER [illegible] FENTON REAGENT AND MICROALGE *CHLORELLA* [illegible] *PSEUDOMONAS FLUORESCENS* BACTERIA 2500, 2000 AND 1500 PPM.

[illegible]	Initial Ratio Analysis FeSO₄ (gram) : H₂O₂				
[illegible]	1 : 2	1 : 4	1 : 6	1 : 8	1 : 10
	8,2	8,3	8,8	8,8	9,7
	22,34	73,12	96,43	103.89	125,78
	1856	1620	1421	1500,03	1823,23
	0,97	0,57	0,56	41,43	103,89
	0,076	0,767	0,230	1,767	6,076
	853,88	746.13	663,56	763.90	986,62
	101	93	100	115	100
	59	59	59	59	59
[illegible]	**1 : 2**	**1 : 4**	**1 : 6**	**1 : 8**	**1 : 10**
	7,8	8,0	8,3	8.7	9.2
	0,375	0,078	0,004	0.0	0,0
	597	520	485	660,65	0.0
	0,73	0,230	0,61	21,23	32.98
	0,062	0,729	0,096	0,729	0,97
	273,995	239,278	226,33	308,30	0,0
	136	99	105	119	114
	60	60	60	69	62
[illegible]	**1 : 2**	**1 : 4**	**1 : 6**	**1 : 8**	**1 : 10**
	7,7	7,9	8,1	8,3	8,7
	0,025	0,003	0,001	0,0 .	0,0
	205	59	0,75	0.0	0,0
	0,26	0,86	0,98	16,98	32,98
	0,056	0,597	0,074	1.597	0,97
	94,32	2.309	0,346	0,0	0,0
	138	101	111	125	114
	62	50	62	62	62

[illegible] source of nitrogen that can be used by the [illegible] nitrate and ammonia-N. The bacteria utilize the [illegible] produced by or derived from dead microalgae [illegible] source for the synthesis of new cells and to [illegible] energy to form the final product such as CO_2, [illegible] the process of respiration and synthesis. [illegible] CO_2 as a carbon source for photosynthesis.

IV. Conclusion

[illegible] ratio between $FeSO_4 : H_2O_2$ in treating waste water [illegible] fertilizer plant using traditional Fenton reagent [illegible] meets the quality standards of waste water quality [illegible] the Minister of Environmental Affairs of the [illegible] of Indonesia No.122 of the year 2004 and the [illegible] of South Sumatra Governor No.18 of of the year [illegible] 1 : 4 , at a concentration of 1500 and 2000 ppm. [illegible] of waste water of urea fertilizer plants by [illegible] advanced oxidation using microalgae *Chlorella* [illegible]*sa*, *Nannochloropsis* sp. *Pseudomonas* [illegible] and the synergy between microalgae *Chlorella* [illegible]*sa* and bacteria *Pseudomonas fluorescen* results [illegible] decrease of pH, NH_3-N, Urea, TKN, Nitrate and [illegible] and an increase of levels of TSS and COD. And [illegible] treatment was using *Pseudomonas fluorescen* [illegible].

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